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CASEFILE

RADIOLYSIS OF WATER IN
SEALED ALUMINUM CAPSULES ITS PREDICTION AND INHIBITION

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RADIOLYSIS OF WATER IN SEALED ALUMINUM CAPSULES ITS PREDICTION AND INHIBITION

by Dean W. Sheibley
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SUMMARY

This report presents the results of an investigation into methods of inhibiting the radiolysis of water in sealed aluminum capsules in the Plum Brook Reactor of NASA.

Experimental design techniques are employed to identify the measured effects of eight variables and their interactions which control water decomposition and the resultant pressure buildup. Multiple regression analyses are used to develop predictive models for the pressure rise observed in an inpile capsule system.

Predictive equations are derived for pressure rise due to the radiolysis of water with air as a cover gas or with helium cover gas containing air as an impurity or mixtures of hydrogen, oxygen, and nitrogen as impurities.

Radiation induced reaction products of nitrogen and oxygen are identified as the key to radiolysis behavior. The prevention of radiolysis depends on the exclusion of air or mixtures of nitrogen and oxygen from the system.

Pure inert gases, helium, neon, and argon prove to be satisfactory radiolysis inhibitors. This is demonstrated for helium by inpile tests lasting up to 14 days (one reactor cycle) in the Plum Brook Reactor.

INTRODUCTION

At the Plum Brook Reactor, the use of water in sealed capsules has created operational problems because of the pressure buildup within these capsules due to the formation of hydrogen and oxygen gases from radiolysis. A search of the literature through 1968 did not present a clear solution to this problem. Therefore, we decided to perform an experiment to identify radiolysis inhibitors and develop predictive equations for the pressure buildup.

This report describes the experiments, the data analysis, and the predictive models. Then the probable cause of radiolysis is discussed and attributed to radiation induced reaction products of nitrogen and oxygen. Finally, proof tests with a pure helium cover gas as a radiolysis inhibitor are described.

DESCRIPTION OF THE EXPERIMENT

A capsule system was designed to simulate the aluminum 'rabbit' capsules which are used for many reactor irradiations. The capsule system consisted of a 6061-T6 aluminum capsule with three delivery lines and two thermocouples. The capsule volume was 28 milliliters and the total system volume was 90 milliliters. System pressure and water temperature were recorded continuously. Gas composition was determined using gas chromatography before and after irradiation. The design of the capsule system hardware (ref. 1) was predicated on the requirements of simplicity and flexibility of operation. Design emphasis was placed on minimizing system volume and on accurate delivery and recovery of test solutions. The nuclear environment was as follows:

Nuclear environment	Range
Gamma heating - W/g Fast flux > 0.1 MeV, neutrons/cm ² -sec Thermal flux, neutrons/cm ² -sec	1.6 to 2.9 2×10 ¹² to 7×10 ¹² 0.95×10 ¹⁴ to 2.9×10 ¹⁴

[This could produce a pressure rise rate of up to 9200 $N/(m)^2$ (sec) (80 psi/hr) based on the G-values reported by Bebesel and Purica (ref. 2).]

Seven variables were tested while the effects of other variables were held constant or minimized. The seven independent variables selected were the following: the water volume; the partial pressures of hydrogen, oxygen, and nitrogen; the reactor power; platinum metal; and cadmium sulfate in solution.

Hydrogen was selected because it is a proved radiolysis inhibitor. Oxygen and nitrogen are the major components of air and in a radiation environment are potentially reactive. Cadmium sulfate was used since it was the most promising soluble inhibitor found in the literature (ref. 3). And platinum metal is a catalyst for hydrogen and oxygen recombination reactions. The capsule water volume and the reactor power are capsule system variables.

These seven variables were controlled at several levels each. In addition, control rod motion in the reactor introduced a measured but noncontrolled eighth variable into

the experiment. The effects of other variables such as water purity, the surface to volume ratio in the capsule, and gas generation from the aluminum-water reaction were minimized either by judicious design or by prior chemical treatment.

In addition to the seven variables, we also evaluated separately the effects of neon, argon, and nitrous oxide. We did not test the effects of krypton or xenon nor the effects of air impurity in neon and argon.

Factorial experiment design techniques were selected for testing the large number of variables. However, due to significant block effects, simple design techniques were not adequate. Therefore, a special ''double telescoping'' blocked experiment design technique was used for the experiment test program. This technique and the detailed statistical design have been previously described in references 4 and 5.

DATA ANALYSIS

Figure 1 shows typical pressure-time behavior of experiment irradiations. The rate of pressure rise normally was linear to approximately 36 hours. The rate then decreased and gradually leveled off. Region A behavior is typical of cover gases of air and of mixtures of hydrogen, oxygen, and nitrogen in helium. Region B typifies behavior of pure helium, hydrogen, oxygen, or nitrogen alone, or mixtures of helium with oxygen, nitrogen, and hydrogen separately.

We chose to predict pressure as a function of time: a linear rate of rise to 36 hours, and nonlinear rise beyond 36 hours. We analyzed the pressure-time data from all test program treatments using a multiple linear regression analysis computer program called RAPIER (ref. 6). A ''backward rejection'' option was used where terms of a predictive model determined insignificant by various statistical tests are deleted

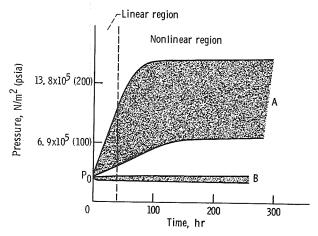


Figure 1. - Typical pressure-time behavior.

sequentially from the model until all terms remaining are acceptable at some preselected level of significance.

Our goal was to derive predictive models for the following conditions:

- (1) Pure helium as a cover gas
- (2) Helium cover gas with air impurity
- (3) Helium cover gas with mixtures of hydrogen, oxygen, and nitrogen
- (4) Air or mixtures of nitrogen and oxygen as the cover gas.

The linear and nonlinear equations to predict pressure are, for irradiations up to 36 hours,

$$P_{t} = P_{0} + a_{0} + a_{1}(\hat{m}t) \tag{1}$$

and, for irradiations between 36 hours and 300 hours,

$$P_{t} = P_{0} + a_{0} + a_{1}(\hat{m}t) + a_{2}(\hat{m}t^{2}) + a_{3}(\hat{m}t^{3})$$
 (2)

where

 P_{t} absolute pressure at time t, psia

 P_0 initial absolute pressure (range, 20 to 40 psia), psia

a constant term from regression analysis (amount by which the equation misses passing through P_0)

 $\mathbf{a}_1, \mathbf{a}_2, \mathbf{a}_3$ regression coefficients

 \hat{m} predicted linear rate of pressure rise up to 36 hr

t length of irradiation

These equations, which predict pressure in psia only, were based on \hat{m} , the linear rate of pressure rise up to 36 hours. The observed rate of pressure rise of experimental treatments was calculated from a least squares fit of pressure-time data taken at 4-hour intervals from 4 to 36 hours. For each study, we derived equations to estimate \hat{m} as a function of the independent variables, their higher degree terms, and interactions. The polynomial equation used to predict \hat{m} is of the form

$$\hat{\mathbf{m}} = \beta_0 + \beta_1 \mathbf{x}_1 + \beta_2 \mathbf{x}_2 + \dots + \beta_i \mathbf{x}_i + \beta_{11} \mathbf{x}_1^2 + \beta_{22} \mathbf{x}_2^2 + \dots + \beta_{ii} \mathbf{x}_i^2 + \beta_{12} \mathbf{x}_1 \mathbf{x}_2 + \beta_{13} \mathbf{x}_1 \mathbf{x}_3 + \dots + \beta_{ij} \mathbf{x}_i \mathbf{x}_j + \dots + \beta_{123} \mathbf{x}_1 \mathbf{x}_2 \mathbf{x}_3 + \dots + \beta_{ijk} \mathbf{x}_i \mathbf{x}_j \mathbf{x}_k + \epsilon$$
(3)

where the x's are the independent variables, and the β 's are the coefficients to be

TABLE I. - RANGES OF VARIABLES FOR VARIOUS

EXPERIMENT PROGRAMS

Reactor control rod bank height, 40 to 60 cm (16 to 30 in.); starting bank height for each irradiation indicated by blocked experiment design.]

Variables	Range	
	Pure helium	
Water ^a volume, ml Initial pressure, N/m ² (p Reactor power, MW	5 to 15 sia) 1. 4×10 ⁵ to 2. 8×10 ⁵ (20 to 40) 40 to 60	
Helium with air impurity		
Water ^a volume, ml Pressure, N/m ² (psia):	5 to 15	
Oxygen	0 to 1. 4×10 ⁴ (0 to 2)	
Nitrogen	0 to 2.8×10 ⁴ (0 to 4)	
Helium	1. 4×10^5 to 2. 8×10^5 (20 to 40)	
Reactor power, MW	40 to 60	
Platinumb	Not present to present	
Helium with hydrogen, oxygen, and nitrogen impurities		
Water ^a volume, ml	5 to 15	
Pressure, N/m ² (psia):		
Hydrogen	0 to 4.2×10 ⁴ (0 to 6)	
Oxygen	0 to 1. 4×10 ⁴ (0 to 2)	
Nitrogen	0 to 5.6×10 ⁴ (0 to 8)	
Helium	1. 4×10^5 to 2. 8×10^5 (20 to 40)	
Reactor power, MW	40 to 60	
Platinum ^b	Not present to present	
Cadmium sulfate ^c	Not present to present	
Air		
Water ^a volume, ml	5 to 15	
Pressure, N/m ² (psia):	4 4	
Oxygen	2.8×10 ⁴ to 5.6×10 ⁴ (4 to 8)	
Nitrogen	1. 12×10^5 to 2. 24×10^5 (16 to 32)	
Reactor power, MW	40 to 60	

^aDeionized water quality: conductivity <1 μ mho/ml; pH 6 to 7. Capsule water temperature range: 328 to 352 K (130° to 170° F.)

bDimensions: area, 5 cm² (0.77 in.²); thickness, 0.0013 cm (0.0005 in.); weight, 0.150 g (3.3×10⁻⁴ lb). cCadmium sulfate solution: 10⁻² molar.

TABLE II. - TERMS AND COEFFICIENTS OF PREDICTIVE MODELS FOR LINEAR RATE OF PRESSURE RISE (EQ. (3))

Regression coefficients	Independent variables	Regression coefficients	Independent variables
Pure helium (12 treatments)		Helium with air impurity (24 treatments)	
There was no significant pressure rise observed. No correlation between variables and observed slopes. Hence, no coefficients and no equation. Helium with hydrogen, oxygen, and nitrogen impurities (44 treatments)		$eta_0 = 19.121$ $eta_3 = -5.312$ $eta_4 = 9.925$ $eta_5 = 2.605$ $eta_6 = -9.989$ $eta_7 = -5.455$	Constant term $x_3(x_3 = 0.667(y_3)-1.000)$ $x_4(x_4 = 0.2(y_4)-1.0000)$ x_5 x_6 x_7
$\beta_0 = 15.782$ $\beta_1 = -0.293$ $\beta_2 = -2.159$ $\beta_3 = 2.734$ $\beta_4 = -1.849$ $\beta_5 = 0.306$ $\beta_6 = -15.150$ $\beta_7 = 0.739$ $\beta_{22} = -2.008$ $\beta_{33} = 2.173$ $\beta_{44} = 5.488$ $\beta_{66} = 3.432$ $\beta_{346} = 3.456$ $\beta_{34/6} = -28.826$ $\beta_{(34/6)(34/6)} = 11.972$ $\beta_{(236)(236)} = -1.732$ $\beta_{23/7} = 11.587$ $\beta_{(23/7)(23/7)} = -7.871$	Constant term $x_1 \\ x_2 \\ x_3(x_3 = 0.667(y_3)-1.0000) \\ x_4(x_4 = 0.2(y_4)-1.0000)$ $x_5 \\ x_6 \\ x_7 \\ (x_2)^2 \\ (x_3)^2 \\ (x_4)^2 \\ (x_6)^2 \\ x_3x_4x_6 \\ x_3x_4/x_6 \\ (x_3x_4/x_6)^2 \\ (x_2x_3x_6)^2 \\ x_2x_3/x_7 \\ (x_2x_3/x_7)^2$	$\beta_{33} = -10.013$ $\beta_{44} = 13.689$ $\beta_{66} = 2.689$ $\beta_{77} = 1.023$ $\beta_{67} = -1.446$ $\beta_{34/7} = -5.157$ $\beta_{55} = 0.424$	$\begin{array}{c} x_{7} \\ (x_{3})^{2} \\ (x_{4})^{2} \\ (x_{6})^{2} \\ (x_{7})^{2} \\ x_{6}x_{7} \\ x_{3}x_{4}/x_{7} \\ (x_{5})^{2} \\ \end{array}$ reatments) $\begin{array}{c} \text{Constant term} \\ x_{3} \\ x_{3} = 0.057 \\ (y_{3}) - 1.17 \\ x_{4} \\ (x_{4} = 0.05 \\ (y_{2}) - 1.00 \\ \end{array}$

Variable	Designation
a ^y 1 a ^y 2 a ^y 3 a ^y 4 y ₅ y ₆ a ^y 7	Water volume, ml Initial hydrogen, N/m ² (psia) Initial oxygen, N/m ² (psia) Initial nitrogen, N/m ² (psia) Reactor power, MW Initial capsule water temperature, K (OF) Initial rod bank height, cm (in.)

Coded variables	Coding constants
x ₁ x ₂ x ₃ x ₄ x ₅ x ₆ x ₇	$0.2(y_1)-2.000$ $0.25(y_2)-1.000$ See individual study See individual study $0.1(y_5)-5.000$ $0.0901(y_6)-28.495$ $0.143(y_7)-1.286$

 $^{^{2}\}text{Coding constants}$ apply to pressure in psia and rod bank height in inches.

estimated. Coefficients with more than one subscript indicate interactions. The ϵ represents the error. The ranges of independent variables are given in table I.

The terms and coefficients of the predictive models for \hat{m} , the linear rate of pressure rise, are given in table II. The numerical values of the regression coefficients are given since they generally indicate the significance and effect on radiolysis. Most variables were coded so that the low level of the range was -1 and the high level was +1. The rod bank height and the absolute temperature were coded between +1 and +3 over the range described in table I. (The air model used a different coding for oxygen and nitrogen to provide a better conditioned matrix. As a result, the algebraic sign of the oxygen and nitrogen coefficients do not directly indicate their effect on radiolysis.)

The coefficients for equations (1) and (2) for predicting pressure are given in table III. The coefficients were derived using the pressure difference from \mathbb{P}_t to \mathbb{P}_0 as the dependent variable and $\hat{\mathbf{m}}$ multiplied by t, t^2 , and t^3 as the independent variable(s). These models predicted the observed values quite well. (The standard error

TABLE III. - TERMS AND COEFFICIENTS OF EQUATIONS (1) AND (2)

	<u> </u>	
	Regression coefficients	
Short term (up to 36 hr) $P_T = P_0 + a_0 + a_1(r)$	nt)	
Helium with air impurity	a ₀ = -0.335 a ₁ = 1.077	
Helium with hydrogen, oxygen, and nitrogen impurities	$a_0 = -1.077$ $a_1 = 0.922$	
Air study	$a_0 = -6.994$ $a_1 = 0.8312$	
Long term (from 36 to 300 hr) $P_T = P_0 + a_0 + a_1(\hat{m}t) + a_2(\hat{m}t^2) + a_3(\hat{m}t^3)$		
Helium with air impurity	$a_0 = -0.605$ $a_1 = 29.725$ $a_2 = -4.527$ $a_3 = 0.213$	
Helium with hydrogen, oxygen, and nitrogen impurities	$a_0 = -1.189$ $a_1 = 25.501$ $a_2 = -3.888$ $a_3 = 0.189$	
Air study	$a_0 = -11.113$ $a_1 = 24.998$ $a_2 = -3.082$ $a_3 = 0.121$	

of the various short and long term models ranged from 5×10^4 to 10×10^4 N/m² (7 to 15 psi).)

The models for predicting the rate of pressure rise behaved well when tested over the range of variables. The size of the standard error was small compared to both computed and observed values. (The standard error of the mean ranged from 25 to 54 newtons per square meter - second (0.23 to 0.47 psi/hr) for the various models.) Based on statistical tests and considerations, these models are statistically significant and are assumed to be valid for use as predictive equations.

DISCUSSION OF RESULTS

Pressure Measurements

The largest linear rate of pressure rise observed in the pure helium study was 12.6 newtons per square meter - second (0.11 psi/hr). There was no correlation between the rates of pressure rise and the independent variables. Therefore, no equation for this case is shown in table II. The small rates of pressure observed probably reflect slight differences in water quality, helium gas purity, and capsule system cleanliness. No significant pressure rise was recorded during a 228-hour irradiation. Area B (fig. 1) indicates typical behavior.

The helium study with air impurity considered only the effects of air (0 to 12 percent by volume) and other mixtures of nitrogen and oxygen in helium. The coefficients in table II indicate that nitrogen promotes radiolysis and the temperature and oxygen inhibit the reactions. (In reality, this effect of nitrogen depends on oxygen also being present.) The rate of pressure rise ranged from 9.9 to 384 newtons per square metersecond (0.086 to 3.34 psi/hr) for 0.4 to 12 percent air in helium. Pure nitrogen (12 percent) in helium and pure oxygen (7 percent) in helium produced no pressure rise in 60 and 20 hour irradiations, respectively. With air impurity, pressure-time behavior generally falls into area A of figure 1.

The mixtures of hydrogen, oxygen, and nitrogen in helium ranged from 0 to 35 percent. Coefficients in table II indicate that temperature has a strong negative effect. Nitrogen and oxygen interacting with temperature also have a strong negative effect. The observed linear slopes had values ranging from 0 to 580 newtons per square metersecond (0 to 5 psi/hr). The slope proved to be very predictable when helium purity was less than 95 percent.

The behavior of all treatments in the air study fall into area A of figure 1. As mentioned earlier, the effects indicated by coefficients in table II are not obvious. The most significant promotor of radiolysis is the high level nitrogen and oxygen interaction.

Oxygen levels greater than 0 (coded) have a strong inhibitive effect. The observed rate of pressure rise ranged from 115 to 590 newtons per square meter - second (1 to 5.1 psi/hr). The longest irradiation in the program lasted 288 hours. Its behavior is indicated well by the upper limit of area A in figure 1.

From results of the regression analysis and comparison of treatment results, cadmium sulfate solution and platinum were without effect in inhibiting radiolysis in capsule system experiments. Capsule system irradiations also showed pure neon and argon to be equally as effective as pure helium in tests up to 60 hours.

Gas Composition Measurements

The most significant observation from the gas composition measurements was the apparent depletion of nitrogen gas. This fact plus the appearance of higher degree terms in the models for oxygen and nitrogen led to an examination of the data to find the source of the radiolysis problem.

It is known (ref. 7) that the irradiation of mixtures of oxygen and nitrogen in a nuclear reactor produces oxides of nitrogen: N_2O , NO, and NO_2 . We also found that the irradiation of either pure nitrogen or pure oxygen produced no gas generation in irradiation tests lasting up to 36 hours.

Several nitrogen oxide formation reactions can be postulated to account for the depletion of nitrogen and the appearance of higher degree terms for oxygen and nitrogen:

$$2N_2 + O_2 = 2N_2O$$
 $K_{eq} = \frac{(N_2O)^2}{(N_2)^2(O_2)}$ (4)

and

$$2N_2O + O_2 = 4NO K_{eq} = \frac{(NO)^4}{(N_2O)^2(O_2)}$$
 (5)

where K_{eq} represents the equilibrium constant for each reaction. The $N_{2}O$ molecule can be drawn in several resonance hybrid forms:

$$: \overset{-}{\ddot{\mathbf{N}}} : \overset{+}{\ddot{\mathbf{N}}} : \overset{+}{\ddot{\mathbf{O}}} : \overset{+}{\mathbf{N}} : \overset{-}{\ddot{\mathbf{O}}} : \overset{+}{\ddot{\mathbf{N}}} : \overset{-}{\ddot{\mathbf{O}}} :$$

The NO molecule may also be considered in valence bond theory as a resonance hybrid (ref. 8):

with the form on the left predominating in the gas phase. The NO molecule is an oddity since it has a free unpaired electron. It is postulated that the $\rm N_2O$ and NO molecules, because of their favorable electronic structures, act as solutes and become involved in the free radical reactions of radiolysis.

Water decomposition by radiation produces both molecular products and free radicals. Fast neutron radiation favors the formation of molecular decomposition products (H_2 and H_2O_2) in addition to the free radical reactions. The latter is related to the presence of gamma radiation and thermal neutrons. Free radical reactions can either promote molecular product formation or enhance recombination. The result of the

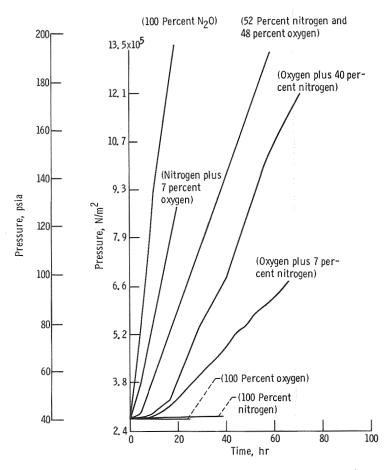


Figure 2. - Measured pressure rise for cover gases of N_2O and for various mixtures of N_2-O_2 .

passage of fast neutrons through water is that groups of free radicals are formed close together along the tracks of the particles. This in effect permits unlike free radicals (H and OH) to immediately recombine to form water. However, the $\rm N_2O$ and/or $\rm NO$ molecules act as solutes in the irradiated water and may significantly hinder the immediate recombination of unlike free radical pairs. The overall effect of these solutes is that of a free radical scavenger, that is, to pick up free radicals. These solutes may even enhance the probability of like radical pair recombination which forms the molecular product hydrogen and, indirectly, oxygen.

The assumption concerning nitrogen concentration and N_2O and NO effects was tested during the course of the experiment treatments. The behavior of these treatments can be seen in figure 2. The most significant treatment is 100 percent N_2O wherein pure N_2O was irradiated. The effect of increasing nitrogen concentration is very apparent. The effect of pure N_2O on the rate of pressure rise is striking evidence of its unquestionable involvement in the radiolysis mechanism. Hence, it appeared that exclusion of mixtures of oxygen and nitrogen should be a satisfactory radiolysis inhibitor for deionized water (no solutes) in aluminum capsules. A test of this theory was made.

PROOF TEST EXPERIMENT

Pure helium was irradiated in aluminum 'rabbits.' The longest irradiation lasted one reactor cycle (14 days). Each rabbit contained 15 milliliters of deionized water and helium cover gas (purity >99.6 percent) at an initial pressure of 3.8×10⁵ newtons per square meter (55 psia). After irradiation, each rabbit was punctured to determine pressure and hydrogen, oxygen, and nitrogen content. Irradiation greater than 64 hours showed a small hydrogen production and pressure increase. Postirradiation pressures ranged from 4.1×10^5 to 4.8×10^5 newtons per square meter (60 to 70 psia). No gas mixture had a flammable composition. This 'fix' was definitely effective.

CONCLUDING REMARKS

It is the opinion of the author that the source of the radiolysis problem in this application is air and the reaction of its radiation induced products with free radicals. The ''fix'' using helium should be equally effective with any container material that does not react chemically with deionized water introducing solutes. Furthermore, the fix should be effective for larger volumes as well. Based on this reasoning, the use of pure helium in a larger inpile aluminum system is planned at the Plum Brook Reactor.

SUMMARY OF RESULTS

The experiment program to investigate radiolysis of water in aluminum capsules produced the following results:

- 1. Predictive equations are given for the rate of pressure rise due to the radiolysis of water in the presence of air, or in the presence of helium cover gas containing air as an impurity or mixtures of hydrogen, oxygen, and nitrogen as impurities.
- 2. Pure inert gases, specifically helium, are shown to be satisfactory radiolysis inhibitors.
- 3. The exclusion of air or mixtures of nitrogen and oxygen will prevent radiolysis of deionized water in aluminum.

Lewis Research Center,

National Aeronautics and Space Administration, Cleveland, Ohio, August 10, 1971, 122-29.

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